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Atmospheric aerosol and ion characteristics during EUCAP (Eucalypt Forest Aerosols and Precursors)

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Abstract. We measured the characteristics and dynamics of atmospheric ions, aerosol particles, and their precursors in an intensive field campaign in a Eucalypt forest in Tumbarumba, South-East Australia, in November 2006. The measured size range of ions was 0.34 to 40 nm and that of aerosol particles approximately 10 to 168 nm, and for observing their size distributions we used an Air Ion Spectrometer (AIS) and a Scanning Mobility Particle Sizer (SMPS). We also measured the hygroscopic and chemical properties of the particles with a Volatility-Humidity Tandem Differential Mobility Analyser (VH-TDMA). The total concentration of ultrafine aerosol particles was measured with a Condensational Particle Counter (CPC). Furthermore, we measured ambient concentrations of volatile organic compounds (VOC), SO_x, NO_x/NO_y, and O_3. Finally, we modelled the 96-h back trajectories of air masses arriving at the site and observed that the arrival directions varied greatly and included trajectories that travelled only over land as well as ones that

travelled most of the time over the ocean. On the most polluted day, the air masses arrived approximately from the direction of greater Sydney / Newcastle coal mine area. The total concentration of ultrafine aerosol particles was approximately 3500 cm^{-3}, and daytime aerosol formation took place on 64% of days with acceptable data**.** The dominant VOCs were isoprene, eucalyptol, a- and b-pinene, camphene, and limonene. The measured hygroscopic growth factors (G_h) at RH of 90% varied from 1.1 to 1.5. The smallest G_h were observed for aged accumulation mode particles in early mornings, and the largest G_h occurred for the freshly nucleated particles on Nov 10, the day with the highest concentration of SO_2.

Key Words: Biogenic aerosol formation; atmospheric ions; VOC; Eucalypt forest; nucleation

INTRODUCTION

Determining the magnitude and driving factors of biogenic aerosol production in different ecosystems is crucial for future development of climate models. So far, most studies of biogenic aerosol production have taken place at continental and coastal sites in the Northern Hemisphere[1]. The EUCAP field campaign in November 2006 was a step towards understanding the process of biogenic aerosol formation from organic vapour precursors in a native Eucalypt forest in South East Australia. Our aim was to determine the characteristics and dynamics of atmospheric ions and charged particles from 0.34 to 168 nm and the composition, concentrations, and fluxes of biogenic volatile organic compounds that react with atmospheric oxidants to form condensable vapours that, in turn, are able to form new aerosol.

MATERIALS AND METHODS

The Tumbarumba flux station is located in a tall open Eucalypt forest in south eastern New South Wales (35° 39' 20.6" S 148° 09' 07.5" E, 1200 m.a.s.l). The dominant species are *E. delegatensis* (Alpine Ash) and *E. dalrympleana* (Mountain Gum), and the average tree height is 40 m. The instrument tower is 70 m tall and it is used to measure fluxes of heat, water vapour and carbon dioxide. Supplementary measurements above the canopy include temperature, humidity, windspeed, wind direction, rainfall, incoming and reflected shortwave radiation and net radiation[2].

We measured the size distribution of air ions (naturally charged clusters and aerosol particles) with an Air Ion Spectrometer (AIS; Airel LTD, Estonia)[3] from 0.34 to 40 nm. The sum of neutral and charged aerosol particles from 10 to 168 nm was measured with a Scanning Mobility Particle Sizer (SMPS, TSI 3936). The volatile and hygroscopic properties were measured with a Volatility-Humidity Tandem Differential Mobility Analyser (VH-TDMA)[4]. The AIS, SMPS, and VH-TDMA were located in a shed on the ground with inlet at the height of 2 m. The total concentration of ultrafine aerosol particles down to ~14 nm was measured with a condensational particle counter (CPC, TSI3010) at the height of 70 m on the tower. Ambient VOCs at the heights of 10 to 50 m were sampled in stainless steel adsorbent tubes filled with Tenax-TA and Carbopack-B to catch both monoterpenes and isoprene and analysed using GC-MS techniques.

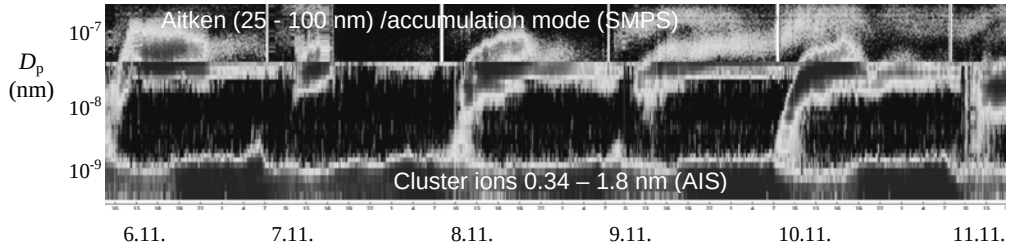

Figure 1. New particle formation measured with an AIS (0.34-40 nm) and an SMPS (figure shows 40-168 nm) during 6. – 11.11.2006. Three normal particle formation events took place on the 6th, 8th, and the 10th of November. Two interrupted events with subsequent Aitken mode growth occurred on the 7th and the 9th. In addition, growth in only the Aitken/accumulation mode was observable on the 9th and 10th. A less clear formation event took place on the 11th. x-axis: time at 3-hour intervals, y-axis: particle diameter. Dark colour in the growth plumes indicates high and light colour indicates low concentration in logarithmic scale. However, the darkest colour on the background indicates very low concentration of particles.

New particle formation was classified as normal (ions grew from clusters all the way to large sizes, Fig. 1); interrupted (ions only grew from cluster to intermediate sizes, Fig.1); Aitken (growth started from intermediate sizes and continued to large sizes, Fig.1)[5]; and nocturnal – sudden nocturnal appearance of large quantities of ions usually in all size classes at the same time but always at least in cluster and intermediate ions (details presented elsewhere[6]).

To determine the source and transport pathways of air masses arriving at Tumbarumba, we analysed back trajectories for the measurement period with the HYSPLIT_4 model, developed by NOAA/ARL[7]. The back trajectories were calculated 96 hours backwards in time at a 70-m arrival height above ground level hourly from 8:00 to 16:00 local time to include the main part of most particle formation events.

RESULTS AND DISCUSSION

Between November 2 and 29, the total concentration of ultrafine aerosol particles was approximately 3500 cm^{-3}. A daytime formation event took place on 18 (64%) of days with acceptable data, and the strongest and clearest event took place on Nov 10, the day with the highest SO_2 concentration (Fig. 1). Nocturnal formation was observable on four nights. Only two days could be classified as entirely aerosol-formation free; the rest of the days were classified as undefined. The air mass arrival directions varied greatly and included trajectories that travelled only over land as well as ones that travelled most of the time over the ocean. On the day with most pollution (Nov 10), the air masses arrived approximately from greater Sydney / Newcastle coal mine area. The dominant VOCs during the campaign were isoprene, eucalyptol, a- and b-pinene, camphene, and limonene. On colder days, also para-cymene became an

important monoterpene. The average midday concentrations at canopy level varied between 11 (b-pinene) and 1011 ng m^{-3} (isoprene).

A detailed discussion on the volatile and hygroscopic behavior of nucleation mode particles is presented elsewhere[8]. In general, the measured hygroscopic growth factors (G_h) at relative humidity of 90% in the VH-TDMA varied from 1.1 to 1.5. The smallest G_h were observed for aged accumulation mode particles measured in the early mornings. The freshly nucleated particles showed G_h varying from 1.15 to 1.5. The largest G_h occurred on Nov 10, the day with the highest concentration of SO_2 (800 pptv as compared to 200 pptv). This is in agreement with previous observations made in a pine forest in Hyytiälä, southern Finland, where a clear correlation between G_h and gaseous H_2SO_4 concentration was observed[9]. Once the nucleation mode has aged and moves into the Aitken mode, the measured hygroscopic growth factor decreases. On Nov 10 for example, G_h decreased from 1.5 to 1.4 in a period of 2 hours. As the particles grow the amount of organic, less hygroscopic material increases, reducing G_h.

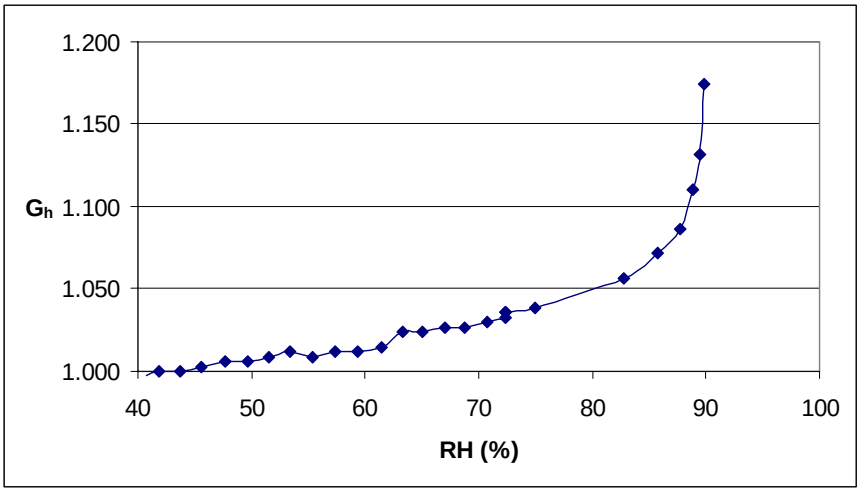

Figure 2. Dependence of the hygroscopic growth factor on relative humidity for a 32-nm particle.

The dependence of G_h on the relative humidity in the VH-TDMA (deliquescence curves) of both nucleation mode particles and accumulation mode particles were also measured. All measured particles showed a continuous growth without exhibiting any deliquescence for RH in the range from 40% to 90%. Figure 2 shows such an example for a 32-nm particle measured several hours after a nucleation event on Nov 8.

Acknowledgments

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